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PHASE MATRIX MEASUREMENTS FOR ELECTROMAGNETIC SCATTERING BY SPHERE AGGREGATES

by

Jerold R. Bottiger, Edward S. Fry, and Randall C. Thompson

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PHASE MATRIX MEASUREMENTS FOR ELECTROMAGNETIC SCATTERING BY SPHERE AGGREGATES

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Abstract

An instrument which permits accurate measurement of all elements of the phase matrix simultaneously has been constructed. Aggregates consisting of attached polystyrene spheres are suspended in an electric field and the phase matrix is measured.

Introduction

During the development of a light scattering photometric polarimeter at our laboratory, numerous scattering matrix measurements were made on aqueous suspensions of nearly monodisperse latex microspheres. Small but significant discrepancies between measured and calculated matrix elements were encountered, most notably in the 2,2 element, which is a sensitive indicator of sphericity. A microscopic examination of the suspensions revealed aggregates of spheres, primarily doublets with decreasing numbers of triplets and higher order aggregates, constituting in total about 10% of the scatterers. The discrepancies were tentatively attributed to the presence of such aggregates.

A study was undertaken to measure directly the scattering matrices of these simple aggregates. The base spheres were 1091 nm diameter (8 nm standard deviation) polystyrene spheres; the light was from a He Cd laser at wavelength 441.6 nm. Results for a doublet, triplet, and quadruplet, as well as the single sphere, are presented below.

The Instrument

The light scattering photometric polarimeter has been described in detail elsewhere.[1,2] Briefly, the device uses four electro-optic modulators (EOM's) to modulate the state of polarization incident upon and scattered from a sample. Figure 1 shows the optical train employed. With a proper choice of the four modulation frequencies and amplitudes, there will exist in the Fourier spectrum of the detected intensity frequencies whose amplitudes are proportional to only one matrix element of the sample. Furthermore, there will exist at least one such frequency for every matrix element, so the entire matrix can be measured

by monitoring amplitudes at appropriate frequencies using standard lock-in techniques. All of the matrix elements are individually measured, simultaneously and in real time, as the detector arm sweeps through scattering angles from ~0° to ~175°.

The dc component of the intensity is proportional to the 1,1 element (phase function) of the sample's scattering matrix. This is held to a constant value, defined to be unity, by a servo system which regulates the high voltage applied to the photomultiplier tube.[3] Thus we actually measure the normalized scattering matrix; the elements are restricted to values between ±1. The phase function itself may be determined either by opening the servo loop or by monitoring the high voltage required in the servo mode and calculating the intensity knowing the photomultiplier tube's response characteristics.

Particle Levitation

To suspend a particle in air, an electrostatic levitation chamber was constructed along the lines described by Fletcher[4] and Wyatt.[5] Figure 2 is a cross-sectional schematic of the apparatus suspending a positively charged particle. Latex spheres, in a solution of water and methanol, are injected by an atomizer into the upper cylindrical chamber. The injection causes the particles to become charged, with approximately equal numbers of positively and negatively charged particles. When the cylinder is rotated to the open position, particles fall through a small hole in the upper plate into the levitation chamber.

The number of spheres in an aggregate is found by clocking its rate of fall in zero electric field. Single polystyrene spheres of 1091 nm diameter were found to require 23 to 27 seconds to fall 1 mm; doublets, 17 to 19 seconds; triplets, 13 to 15 seconds; and quadruplets, 11 to 12 seconds. The spread in velocity for

a particular particle results from Brownian motion. It is difficult to distinguish among aggregates of more than five because of overlapping velocity ranges.

Once a particle is selected, the top cylinder is rotated closed and the particle is held midway between the top and bottom plates by manually adjusting the potential between the plates. The inhomogeneous field, which provides a radially inward force centering the desired particle horizontally, also drives out all other particles within a minute or two. Finally, a servo system is activated which optically senses the particle's vertical position and adjusts the plate potential as needed to keep the particle at a constant height.

Although trapped in a very small volume, the particle is still subject to Brownian motion and rapidly changes its orientation. This is clearly observed as a scintillation of the intensity, except in the case of a single sphere where the intensity is quite steady. So, while the sample is a single particle, the scattering matrix measured is equivalent to that of a cloud of identical particles in many orientations.

Results

Figure 3 shows the 1,2 scattering matrix element for a singlet, doublet, triplet, and quadruplet of 1091 nm spheres for scattering angles from 12° to 165°. (This somewhat restricted range of scattering angles was necessitated by the design of the electrostatic levitation chamber requiring very low levels of stray light.) The single particle Mie structure is clearly evident in the scattering of the aggregates, although it becomes increasingly washed out as the aggregate grows in complexity. The same behavior is seen in the 4,3 elements, Figure 4.

For all four samples, the 2,1 and (-)3,4 elements were identical to the corresponding 1,2 and 4,3 elements and are not shown. The eight elements which

vanish for spheres (1,3; 1,4; 2,3; 2,4; 3,1; 3,2; 4,1; 4,2) were measured and found to be zero for all aggregates as well as the single sphere. These also are not presented. The vanishing of these eight elements for the aggregates is consistant with the interpretation that we have in effect measured the particles averaged over all orientations.

The 2,2 matrix element (Figure 5), which is unity at all scattering angles for a particle with spherical symmetry, drops strikingly in the case of aggregates of spheres. This element provides the most unequivocable evidence for the non-sphericity of the aggregates.

The 3,3 and 4,4 matrix elements are presented in Figures 6 and 7. They should be, and are found to be, the same for a single sphere. However, even with random orientation there is no a priori reason to expect them to agree in the case of aggregates. In fact, they are not identical, as is most clearly seen in the backward scattering angles of the quadruplet.

Conclusion

Depending upon which matrix elements of the aggregates are examined, we can observe evidence of both the non-sphericity of the overall aggregate structure (eg, the 2,2 element) and the monodisperse spherical substructures. We feel it is important, especially when attempting to characterize irregular particles by their light scattering properties, that the entire scattering matrix be determined.

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Fig. 1. Schematic diagram of the photometric polarimeter.

Fig. 2. Schematic diagram of the particle levitation chamber.

Fig. 3. 1,2 element of the scattering matrices for single and aggregate spheres.

Fig. 4. 4,3 element of the scattering matrices for single and aggregate spheres.

Fig.5. 2,2 element of the scattering matrices for single and aggregate spheres.

Fig. 6. 3,3 element of the scattering matrices for single and aggregate spheres.

Fig. 7. 4,4 element of the scattering matrices for single and aggregate spheres.

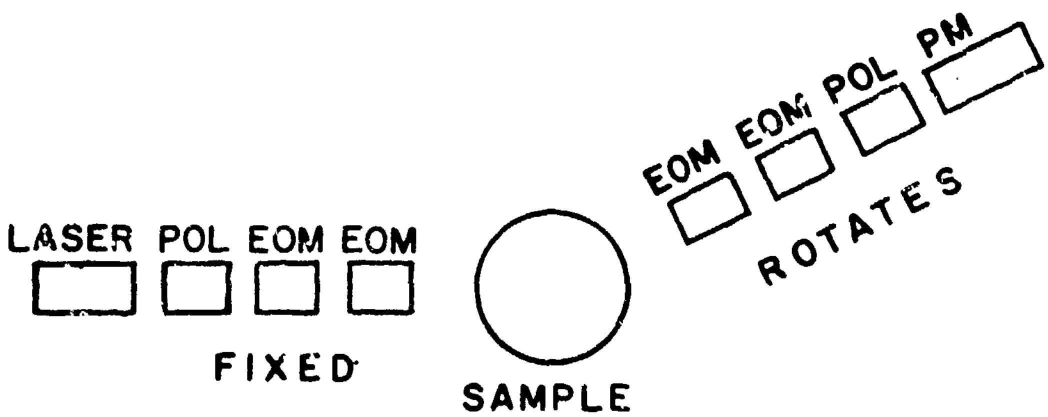

Fig. 1. Schematic diagram of the photometric polarimeter.

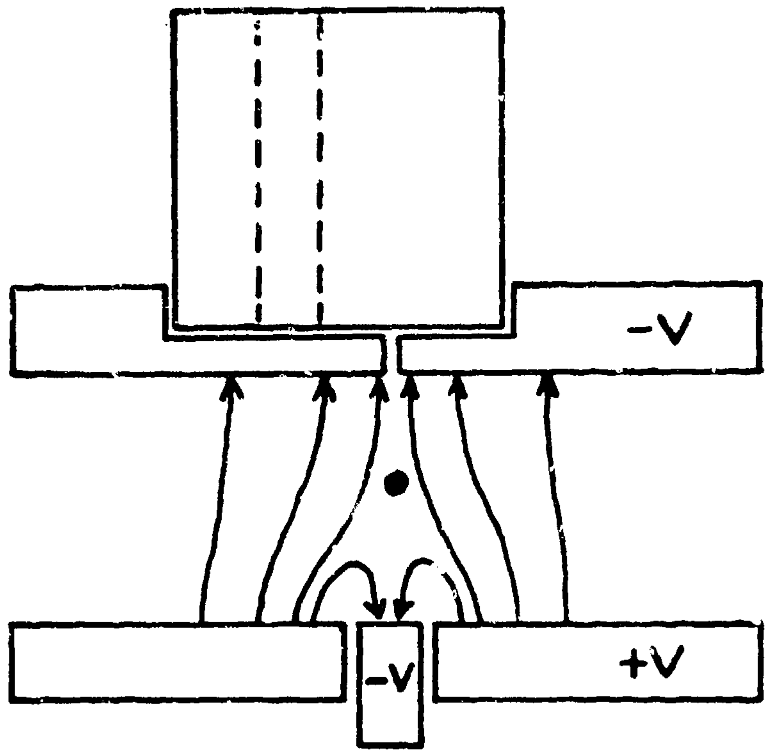

Fig. 2. Schematic diagram of the particle levitation chamber.

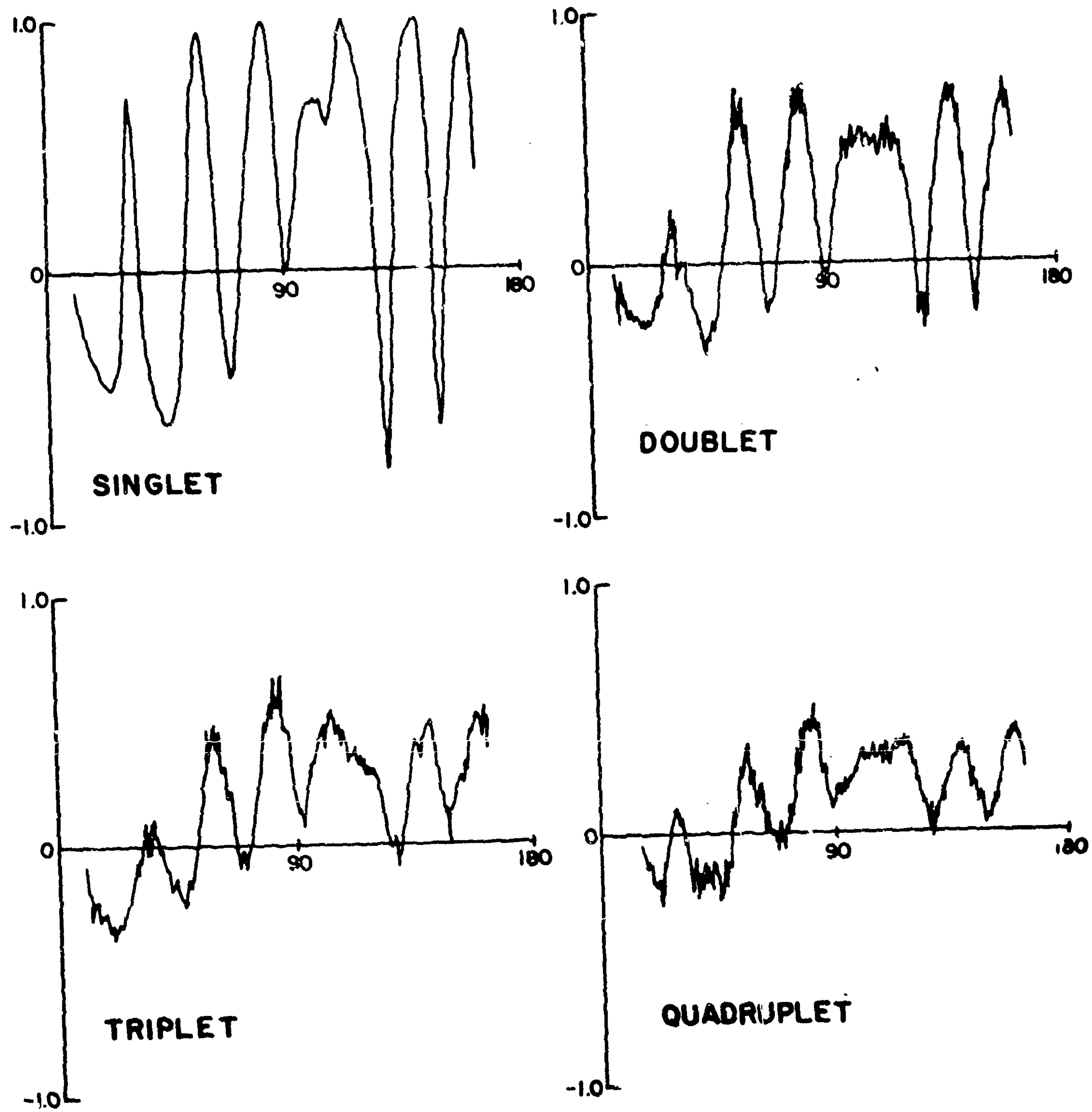

Fig. 3. 1,2 element of the scattering matrices for single and aggregate spheres.

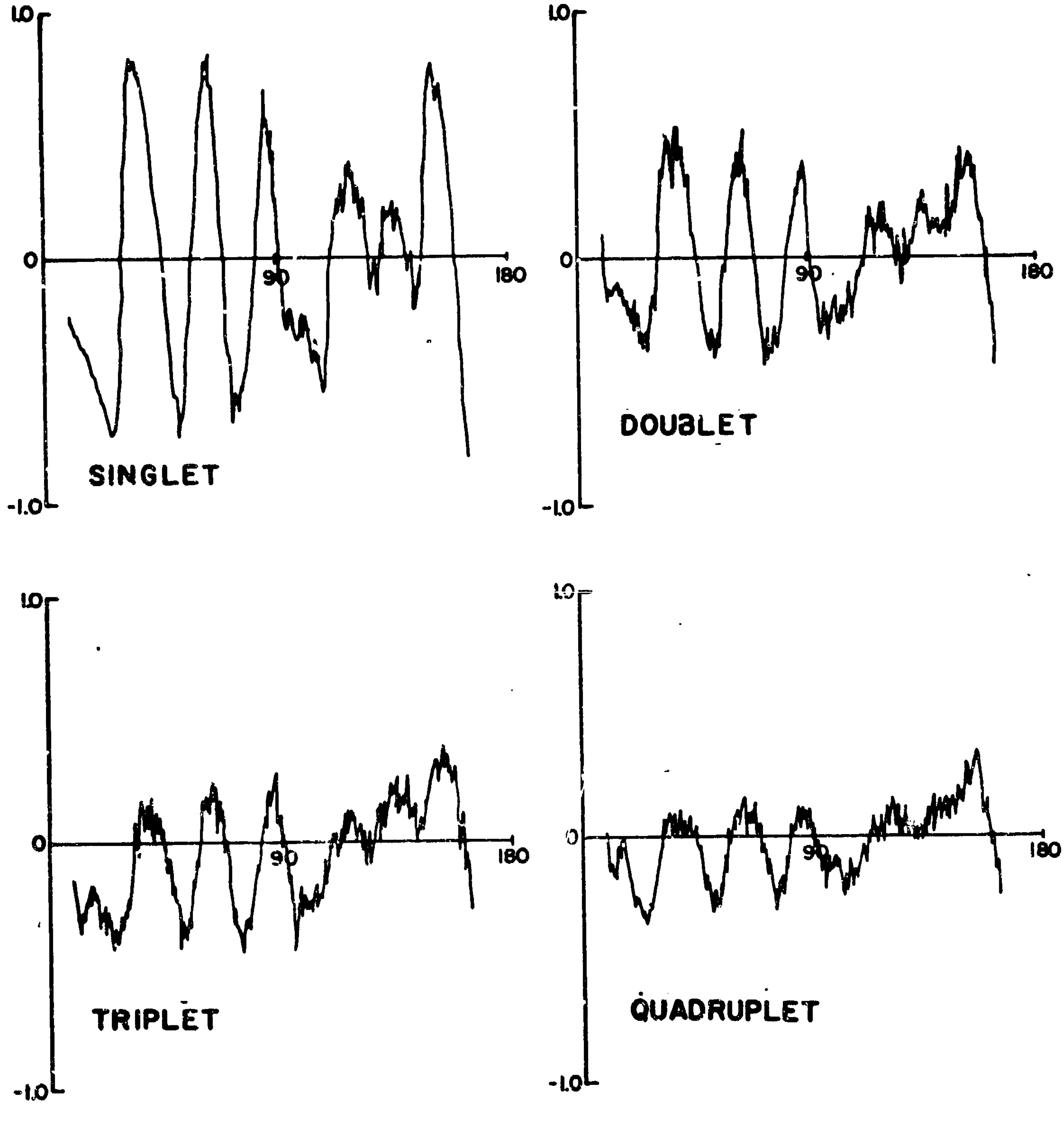

Fig. 4. 4,3 element of the scattering matrices for single and aggregate spheres.

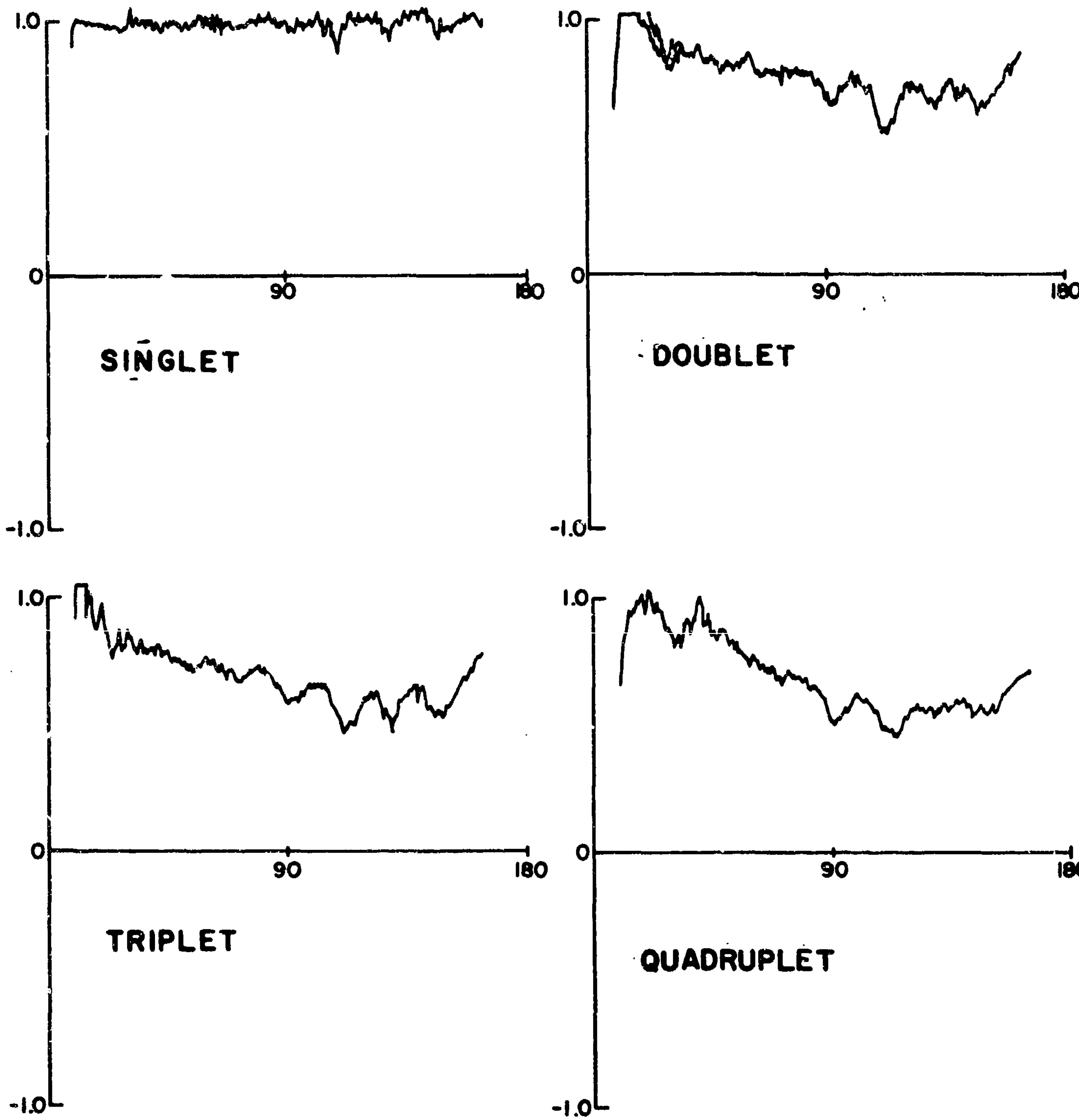

Fig.5. 2,2 element of the scattering matrices for single and aggregate spheres.

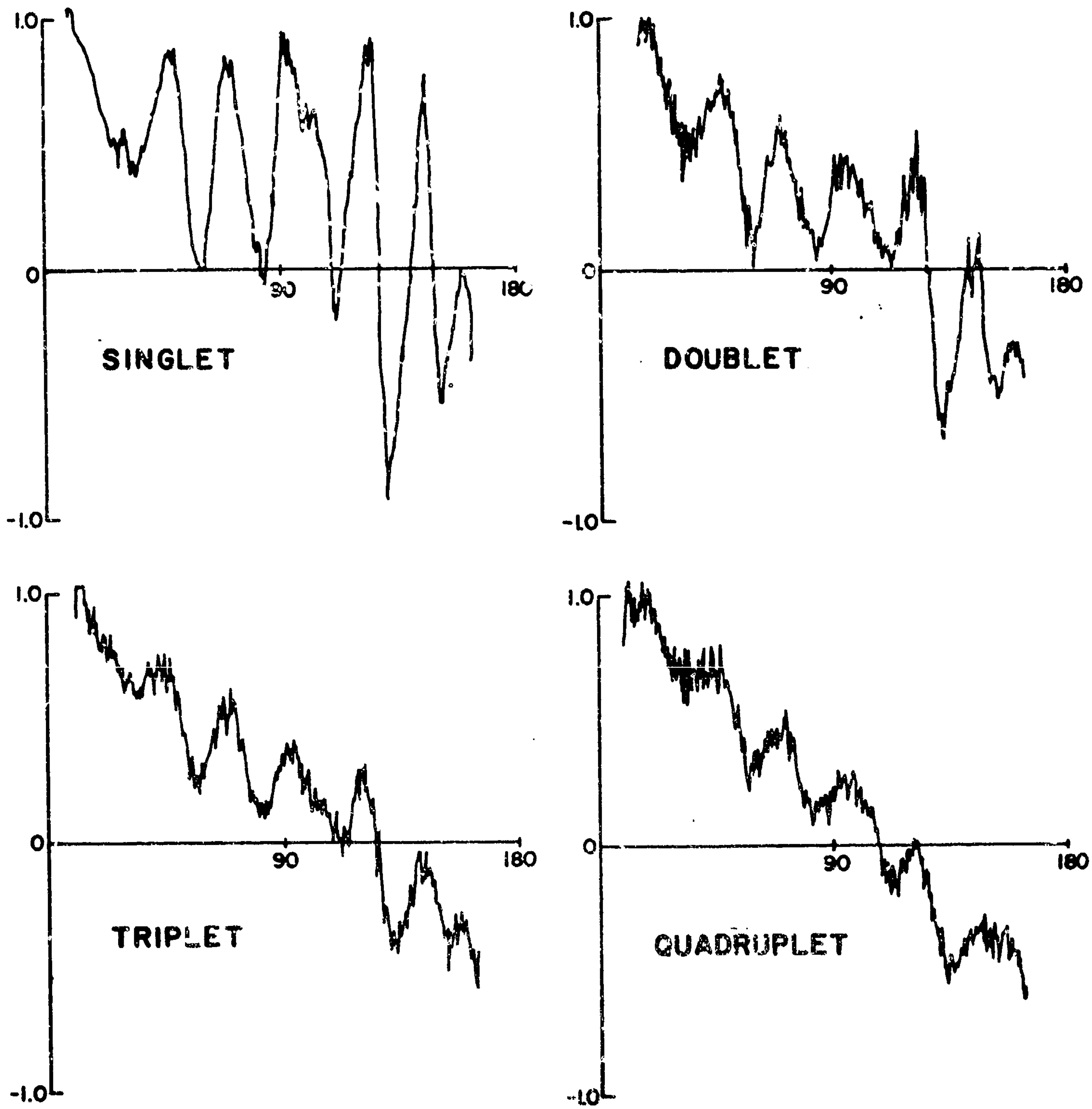

Fig. 6. 3,3 element of the scattering matrices for single and aggregate spheres.

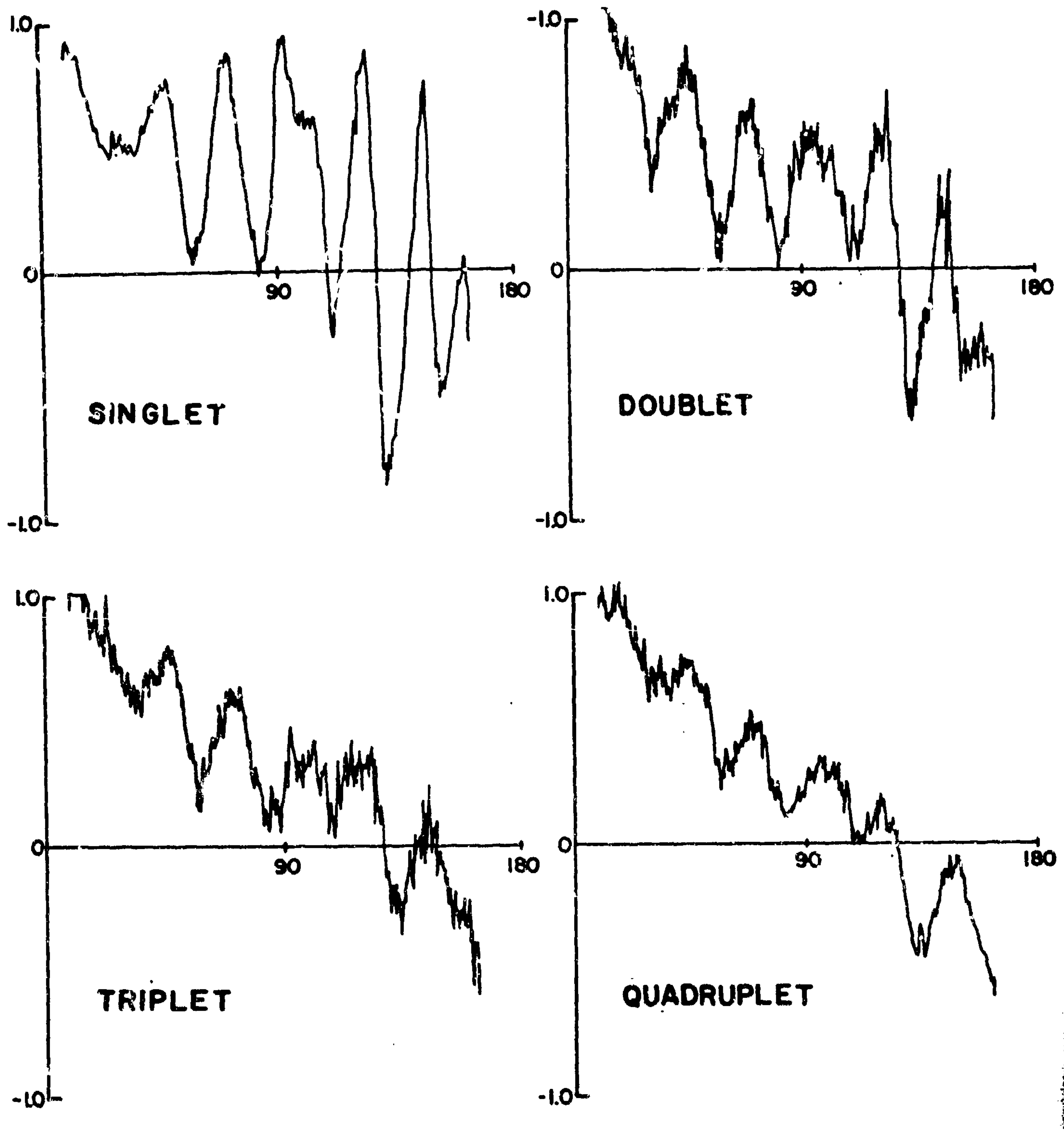

Fig. 7. 4,4 element of the scattering matrices for single and aggregate spheres.